Probing Auger decays in the time domain

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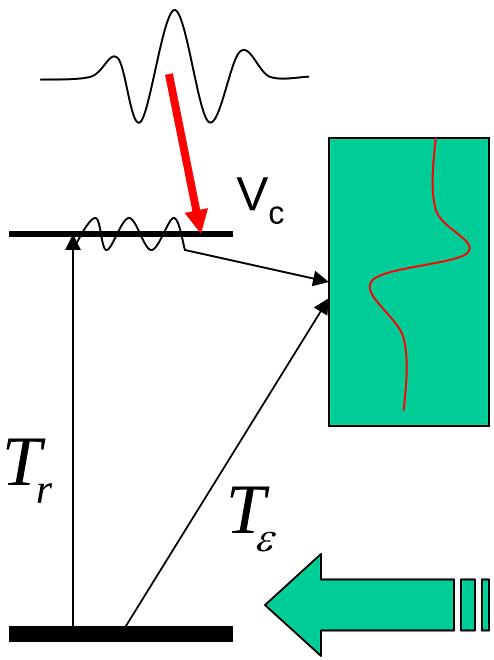
1. **Energy domain vs time domain Measurements**
2. **X-ray Pump- IR laser probe on electron dynamics**
3. **Theory of Laser-assisted autoionization (Auger decay)**
4. **AC Stark effect in laser-assisted x-ray photoionization**
5. **Opportunities with picosecond x-ray pulses**

1. Energy Domain description

an autoionizing state is described by Fano's formula



$$f_F(E) = \frac{q+\epsilon}{1-i\epsilon}$$

Reduced energy: $$\epsilon = \frac{E - E_\phi - F(E)}{\frac{1}{2}\Gamma}$$

Shifted resonance position: $$E_r = E_\phi - F(E)$$

Resonance width: $$\Gamma$$

q parameter: . $$q = \frac{T_r}{\pi V_c^* T_\varepsilon}$$

The lifetime is deduced from $1/\Gamma$ -- about 1-10 fs

What is needed in the time-domain measurements?

start

a few fs

stop

Pump

probe

Current technology:

Weak XUV pulses -- 0.5fs (thru Harmonic Generation),

about 100 eV energy,

Intense IR laser -- 5fs, 780nm mean wavelength,

few cycles; carrier phase

atoms

rather well understood in energy domain

Time-resolved measurements: previous work

With:

attosecond soft-X-ray and fs laser pulse,

- Cross-correlation can be built for laser assisted photoionization to:
  - Measure X-ray pulse duration[1,2]
  - Measure absolute phase of the laser pulse(?)
- Measure the lifetime of a resonance: laser assisted auger decay [3]
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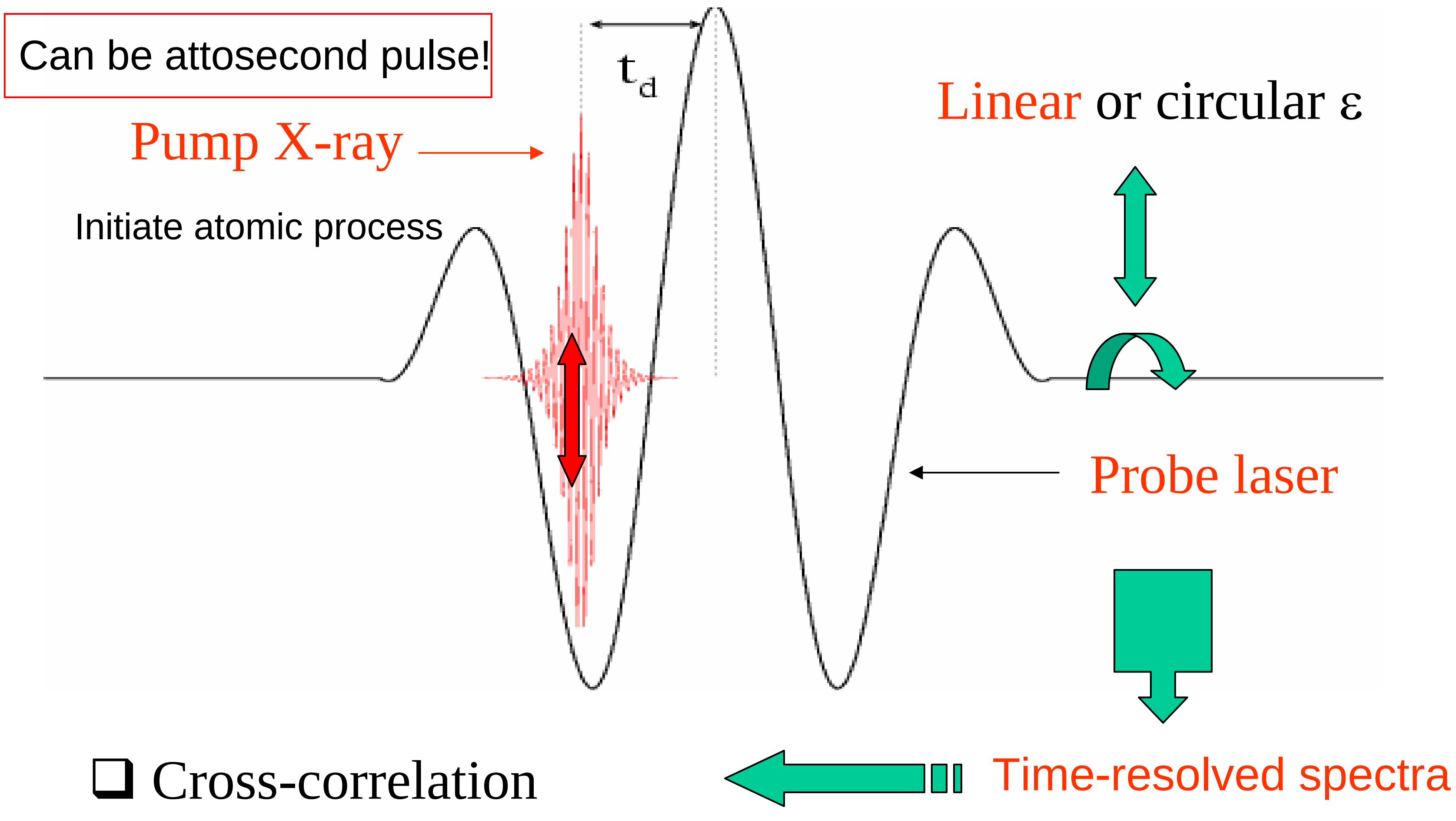
1. Hentschel et al, Nature 414, 509
2. Drescher et al, Science 291, 1923
3. Drescher et al, Nature 419, 803

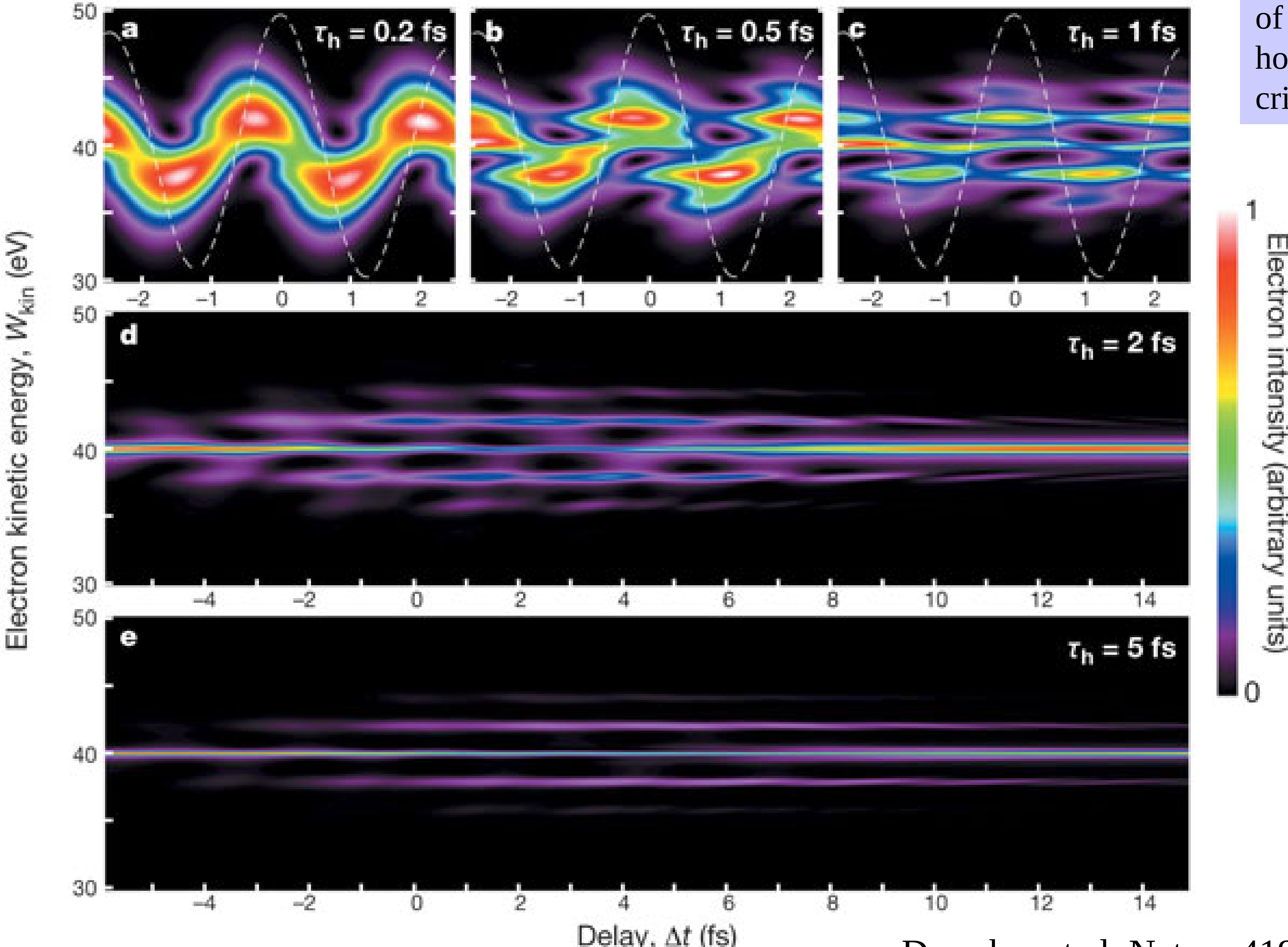
Illustration of pump-probe schemes



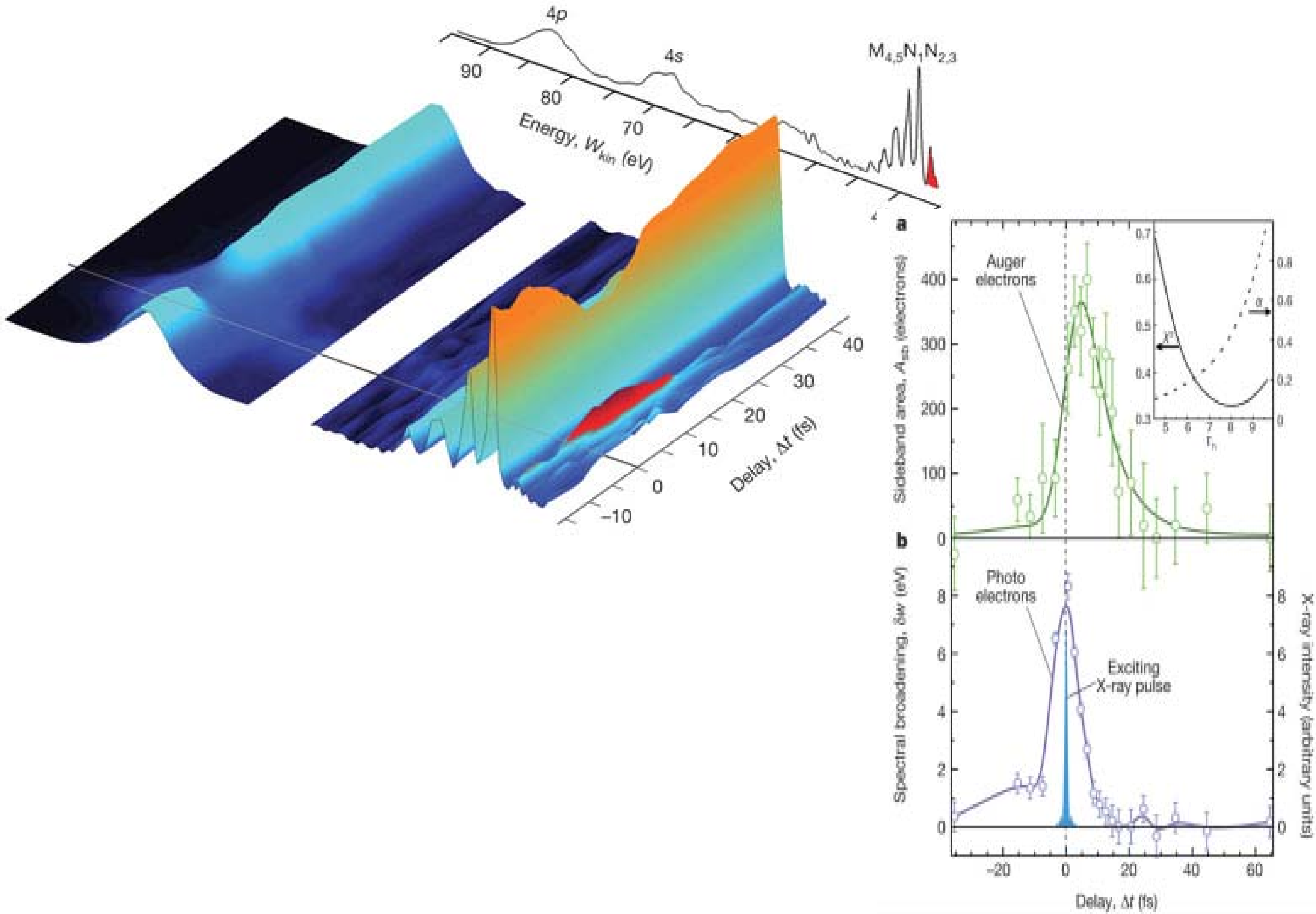
- Cross-correlation
- Probe atomic dynamics

Photoelectron spectra vs time delay—from model

The liftetime of the hole is critical



Drescher et al, Nature 419, 803



4p
4s
$M_{4,5}N_1N_{2,3}$
90
80
70
Energy, W_{kin} (eV)
Delay, Δt (fs)
40
30
20
10
0
−10
a
Sideband area, A_{sb} (electrons)
Auger electrons
b
Spectral broadening, δw (eV)
X-ray intensity (arbitrary units)
Photo electrons
Exciting X-ray pulse
Delay, Δt (fs)
τ_h

Our work—

1. To present a rigorous and simple theory for obtaing the electron spectra for such pump-probe experiments → to extract the lifetime of an Auger resonance: **to be used for probing the parameter space by experimentalists**

1. To generalize the theory to autoionization states
2. To generalize to more than one resonance states

submitted to PRL

Formulation of Laser-assisted PI

Assumptions

Free electron: ~~Coulomb field~~, laser field, ~~X-ray field~~

Strong field approximation

$$\Psi = |0\rangle \exp(iI_p t) + \int d^3 p b(\vec{p},t) |\vec{p}\rangle$$

Bound electron: ~~excitation~~

~~depletion of ground state~~

Photoionization: ~~Laser field~~, X-ray field

Electron amplitude:

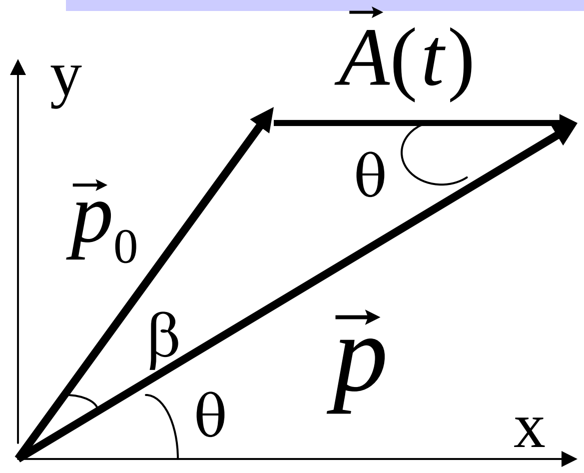
$$b(\vec{p}) = i \int_{-\infty}^{\infty} dt' \vec{E}(t') \cdot \vec{d}[\vec{p} - \vec{A}(t')]$$

$$\times \exp\{-i \int_{t'}^{\infty} \frac{[\vec{p} - \vec{A}(t")]^2}{2} dt'' + iI_P t'\}$$

Stationary phase equation: $\frac{1}{2}[\vec{p} - \vec{A}_l(t_s)]^2 = W_0$ t_s: Saddle point

$$W_0 = \hbar\omega_x - I_p$$

Kinematics



$$\frac{1}{2}[\vec{p}-\vec{A}(t)]^2 = W_0$$:energy conservation

Linear polarization: $\vec{A} = A\sin(\omega t)\vec{e}_x$

Electron energy at observation angle θ:

$$W_p = W_0 + 2U_p(t)\sin^2\omega_l t\cos 2\theta + \cos\beta\sqrt{8W_0U_p(t)}\sin\omega_l t\cos\theta$$

$$\cos\beta = \pm\sqrt{1-2U_p\sin^2\theta\sin^2\omega_l t/W_0}$$

Or:

$$W_p \approx W_0 + U_p(t)(\cos 2\omega_l t - 1) + 4U_p(t)\sin^2\omega_l t\cos^2\theta + \sqrt{8W_0U_p(t)}\sin\omega_l t\cos\theta$$

Laser-assisted autoionization: Lorentzian shape

Field-free:

Time profile: $f(t) = \exp(-iE_r t - \frac{\Gamma}{2} t)$

Energy domain: $f(E) = \int dt f(t) \exp(iEt) = -\frac{1}{i(E - E_r) - \Gamma/2}$

Laser-assisted: electron spectrum under strong field approximation (SFA):

$$\begin{aligned} b_L(\vec{p}, t) &= i \int_{-\infty}^{t} dt_1 \int_{t_1}^{t} dt_2 \vec{E}(t_1) \cdot \vec{d}(\vec{p}) e^{iI_p t_1} & (1) \\ &\times \exp[(-iE_r - \frac{\Gamma}{2})(t_2 - t_1)] & (2) \\ &\times \exp\{-i \int_{t_2}^{t} \frac{[\vec{p} + \vec{A}(t) - \vec{A}(t_3)]^2}{2} dt_3\} & (3) \end{aligned}$$

Virtual three-step process:

1. Resonance state excited by X-ray at time t_1;
2. Decay at time t_2 giving birth to continuum electrons;
3. Propagation of electrons in the laser field.

Laser-assisted autoionization: Fano shape

Profile in energy domain:
$$f_F(E) = \frac{q+\epsilon}{1-i\epsilon}$$

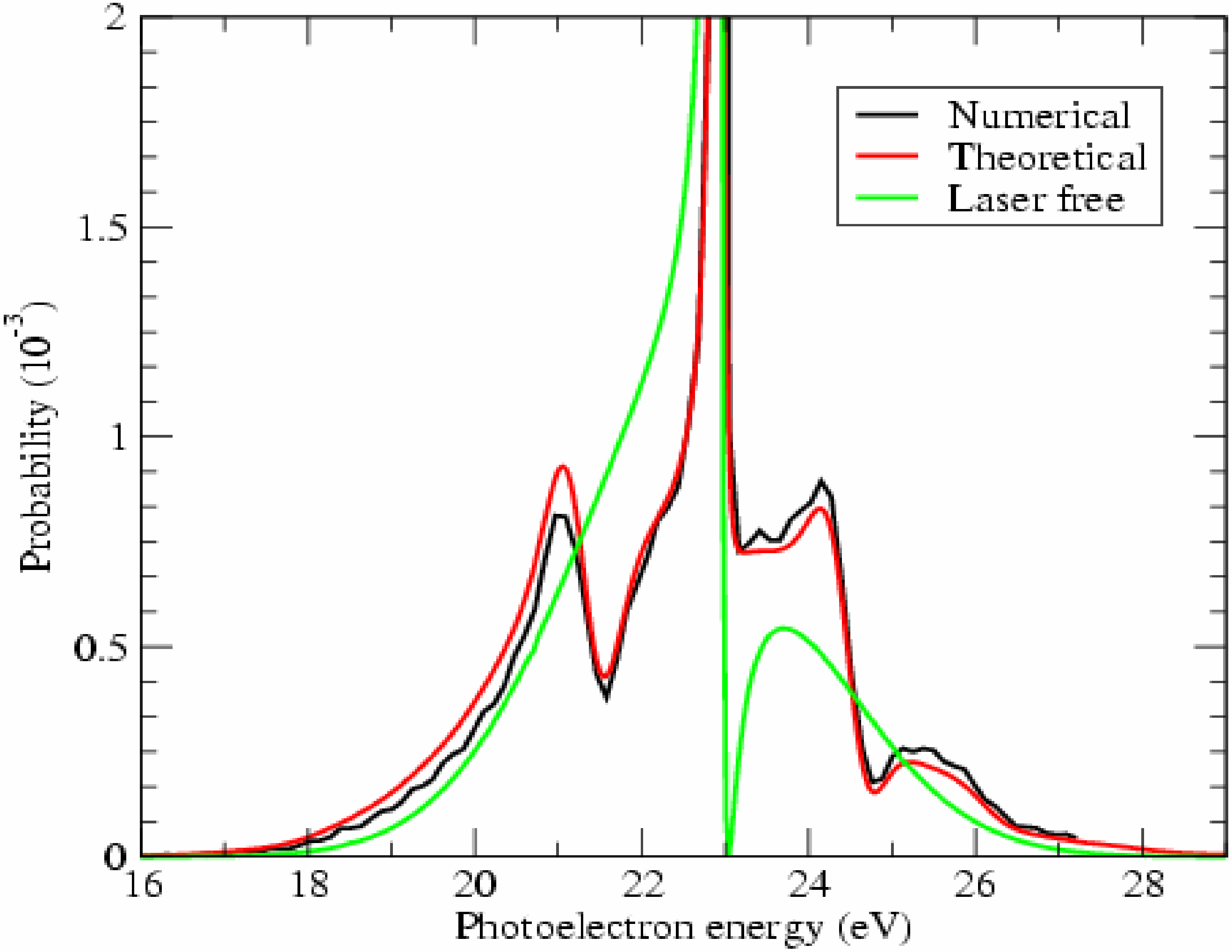
Profile in time domain:
$$\boxed{F_F(t) = \frac{\Gamma}{2}(q-i)e^{-iE_\phi t-\Gamma t/2} + i\delta(t-0)}$$

$$\begin{aligned} b_F(\vec{p},t) &= i\int_{-\infty}^{t} dt_1 \int_{t_1}^{t} dt_2 \vec{E}_x(t_1)\cdot\vec{d}(\vec{p}')e^{iI_p t_1} && (1)\\ &\times \boxed{[\frac{\Gamma}{2}(q-i)e^{(-iE_r-\frac{\Gamma}{2})(t_2-t_1)} + i\delta(t_2-t_1)](2)} \\ &\times \exp\{-i\int_{t_2}^{t}\frac{[\vec{p}+\vec{A}(t)-\vec{A}(t_3)]^2}{2}dt_3\} && (3)\end{aligned}$$

$$\Gamma, E_\phi, q \rightleftarrows b_L(\vec{p},t)$$

?

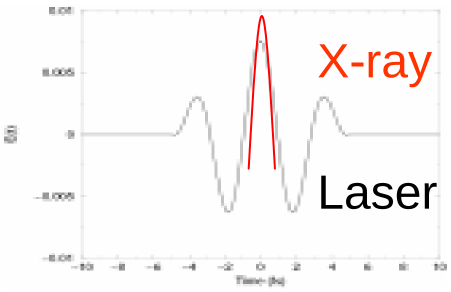
Angle-Integrated spectra (delay= 0)



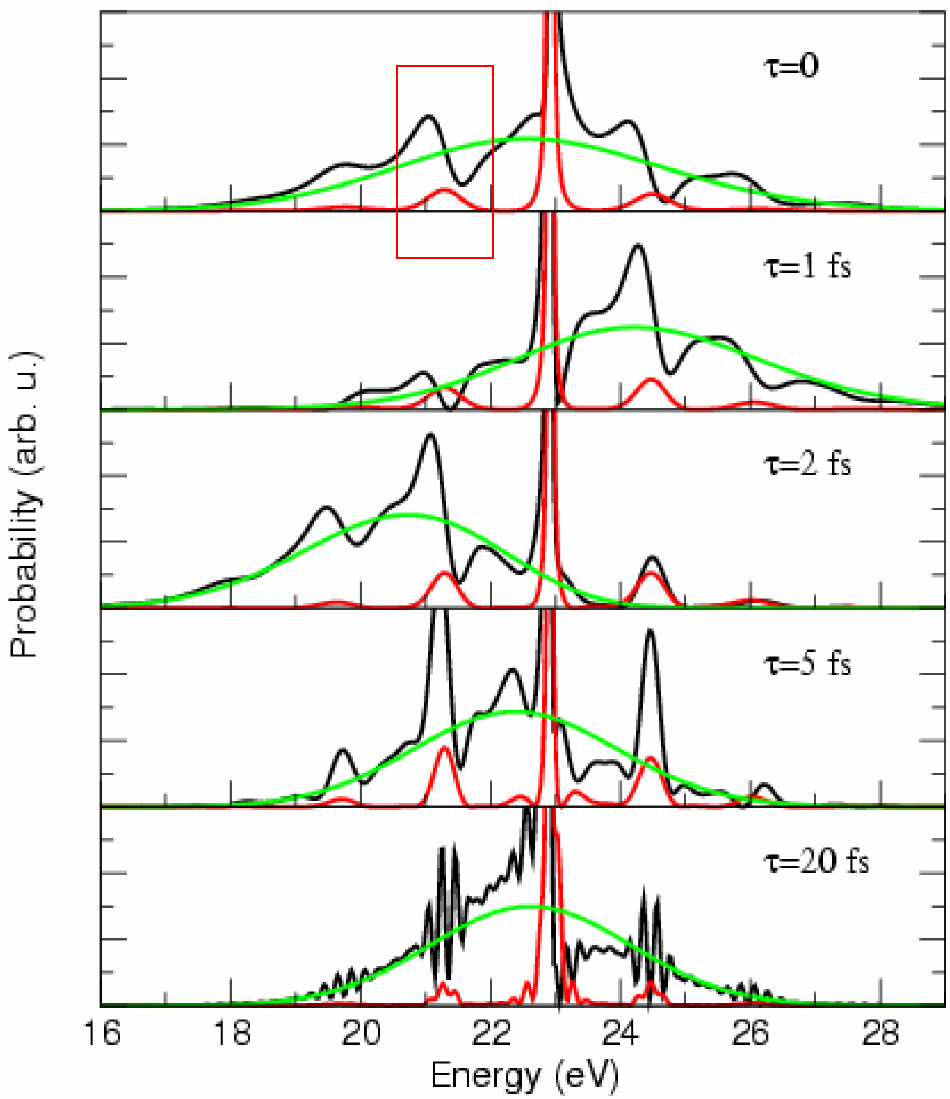
Fano resonance: 22.9 eV (position), 0.055 eV (12 fs) (width) and -4.2 (q number).

Xray: 0.5 fs, 1×10^{12} W/cm^2, 39 eV

Laser: 5 fs, 1×10^{12} W/cm^2. Phase:0 and frequency 1.65 eV (~759nm,2.5fs).



Electron spectra in the forward direction vs time delay



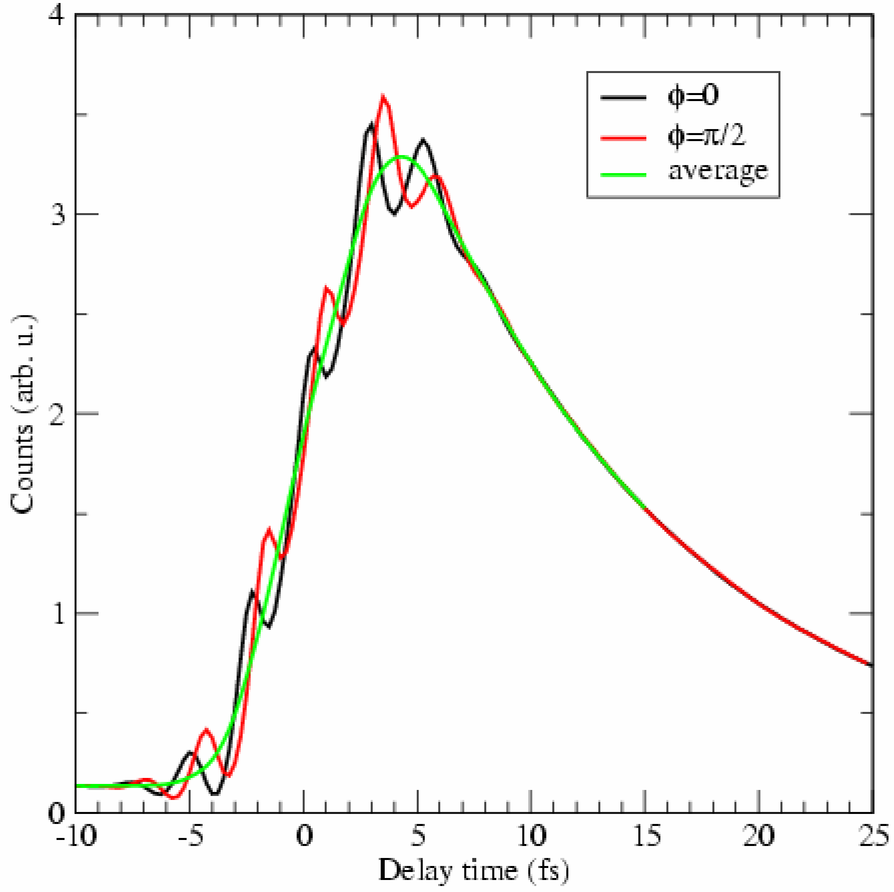
direct

“bound” only

Total

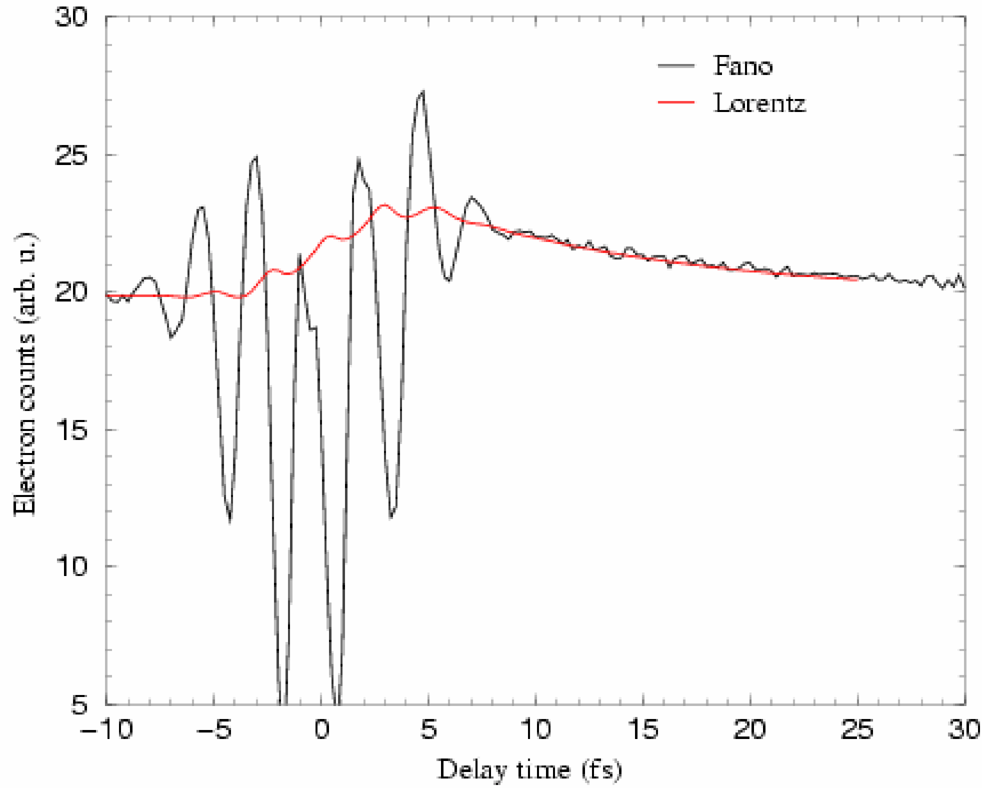
$$\Delta\phi = (E - E_r)t_d$$

Determine lifetime of a Lorentzian resonance



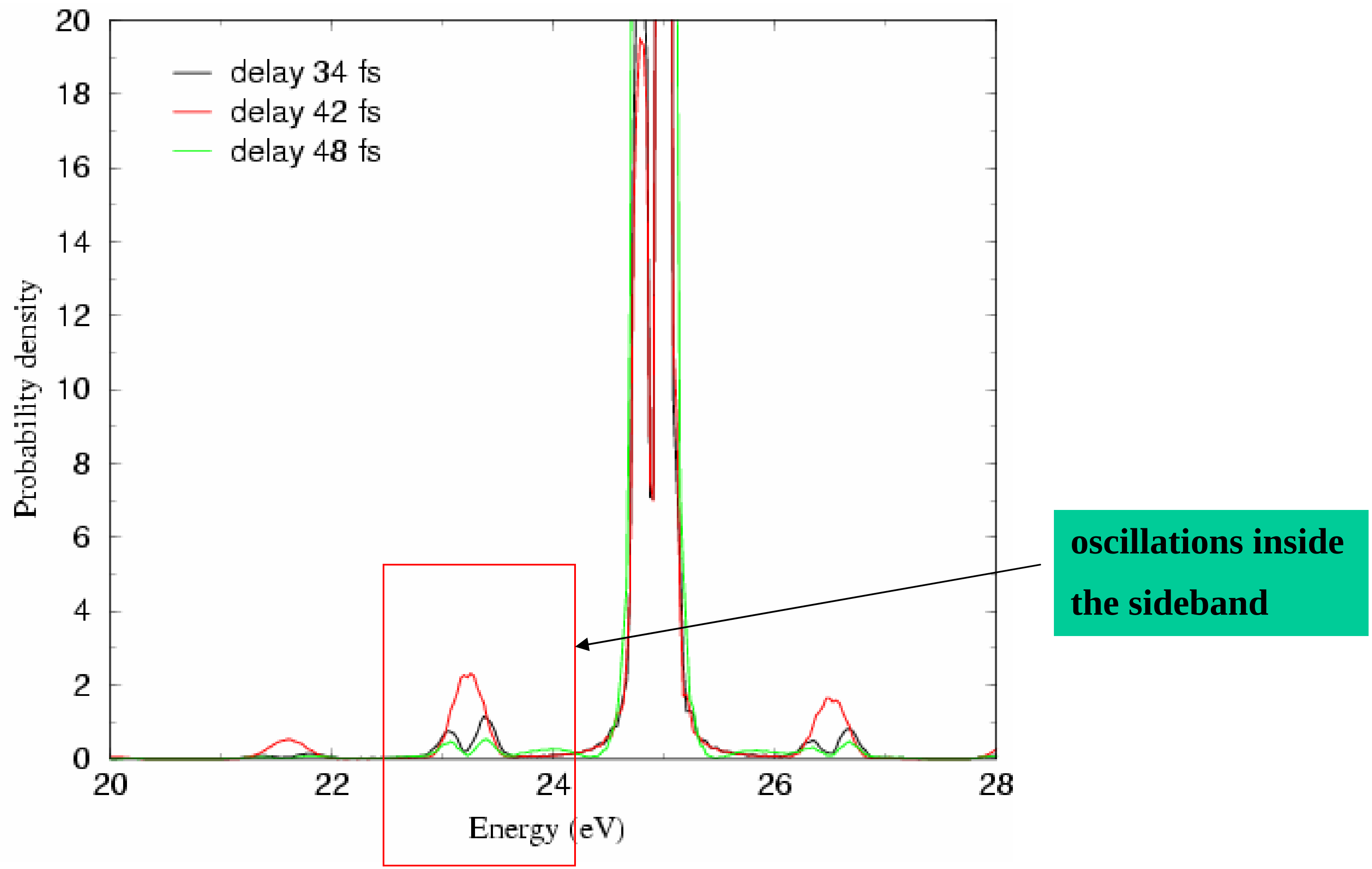
Electron counts within first sideband (20.4-22.1 eV) verse time delay between two pulses for given laser Int. but different laser carrier phases.

Determine lifetime of a Fano resonance



Electron counts within first sideband (20.4-22.1 eV)
verse time delay between two pulses

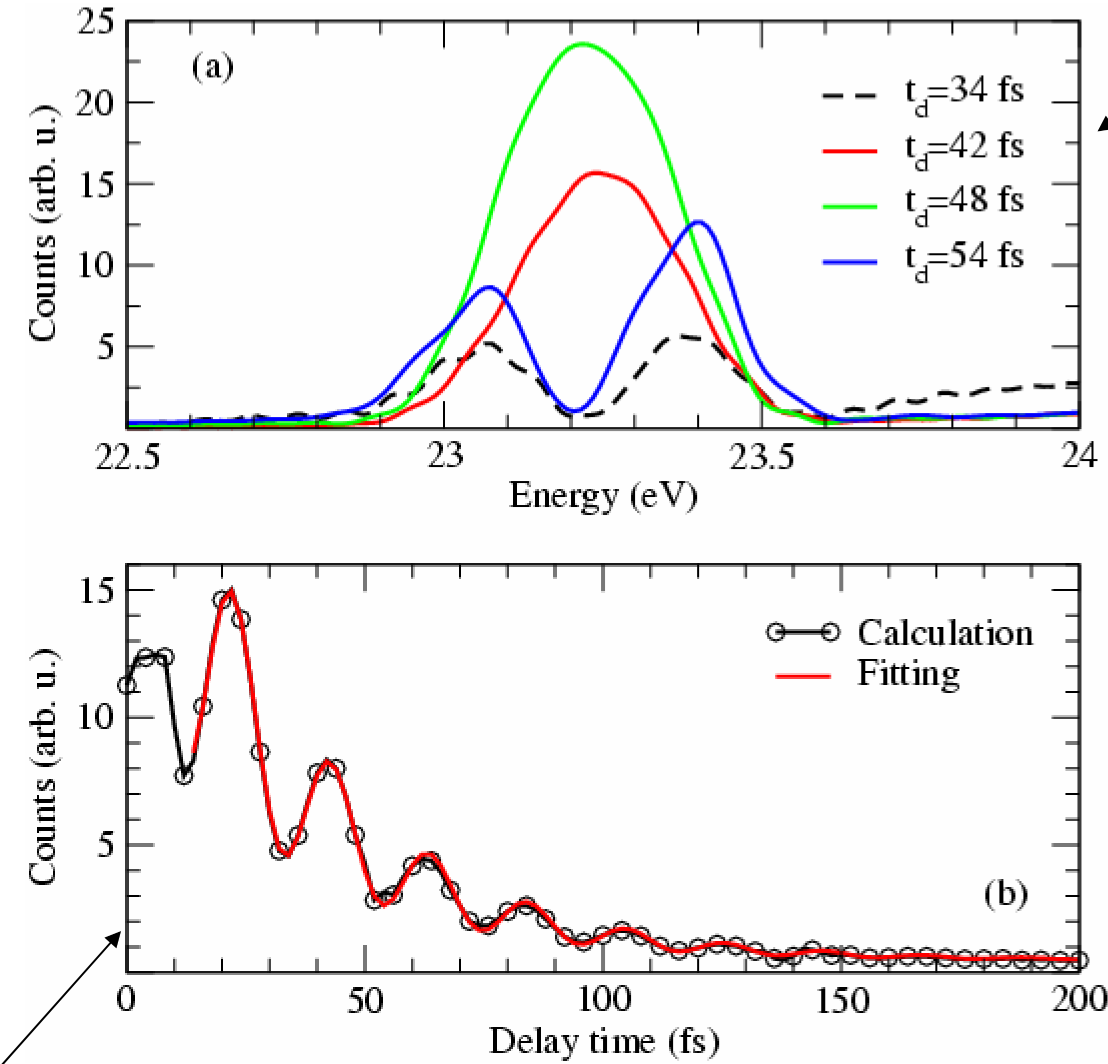
Two nearby Lorentz resonances in a laser field



Energy positions (lifetimes) of two resonances:24.8 eV (30 fs), 25 eV (40 fs)
XUV duration 0.5 fs with freq. 41 eV, Laser duration 10 fs with freq. 1.65 eV.
Intensities of Laser and XUV pulse are both 10^{12}W/cm^2.

Measuring energy separation from quantum beat

spectra at the first sideband



Phase difference: $(E_2\text{-}E_1)t_{delay}$
👎E=0.2 EV correspondent to 21 fs

Two resonances:24.8 25 eV with lifetime 30 and 40 fs. Laser duration 10 fs with freq 1.65 eV, X-ray 0.5 fs. Both intensity 10^12 W/cm^2

area under the first sideband

Fitted by: $$c_1 e^{-\Gamma_1 t} + c_2 e^{-\Gamma_2 t} + c_{12} e^{-(\Gamma_1+\Gamma_2)t/2} \cos(\Delta E t + \phi_0)$$

AC stark effect by the laser on the photo-ionization of helium by short XUV light pulses

1. Effect of drift velocity– as in strong field approximation
2. **Stark effect – electric field induced new resonances -- new**
3. **Multiphoton absorption of laser photons-- new**

→ Need to solve:

time-dependent Schrodinger equation for two-electron atoms

Application: He** in a Static Field

$$He(1s^2\ {}^1S) + h\nu \rightarrow He^{**}(2l\ nl'\ {}^1P)$$

(to dipole allowed states)

Static field

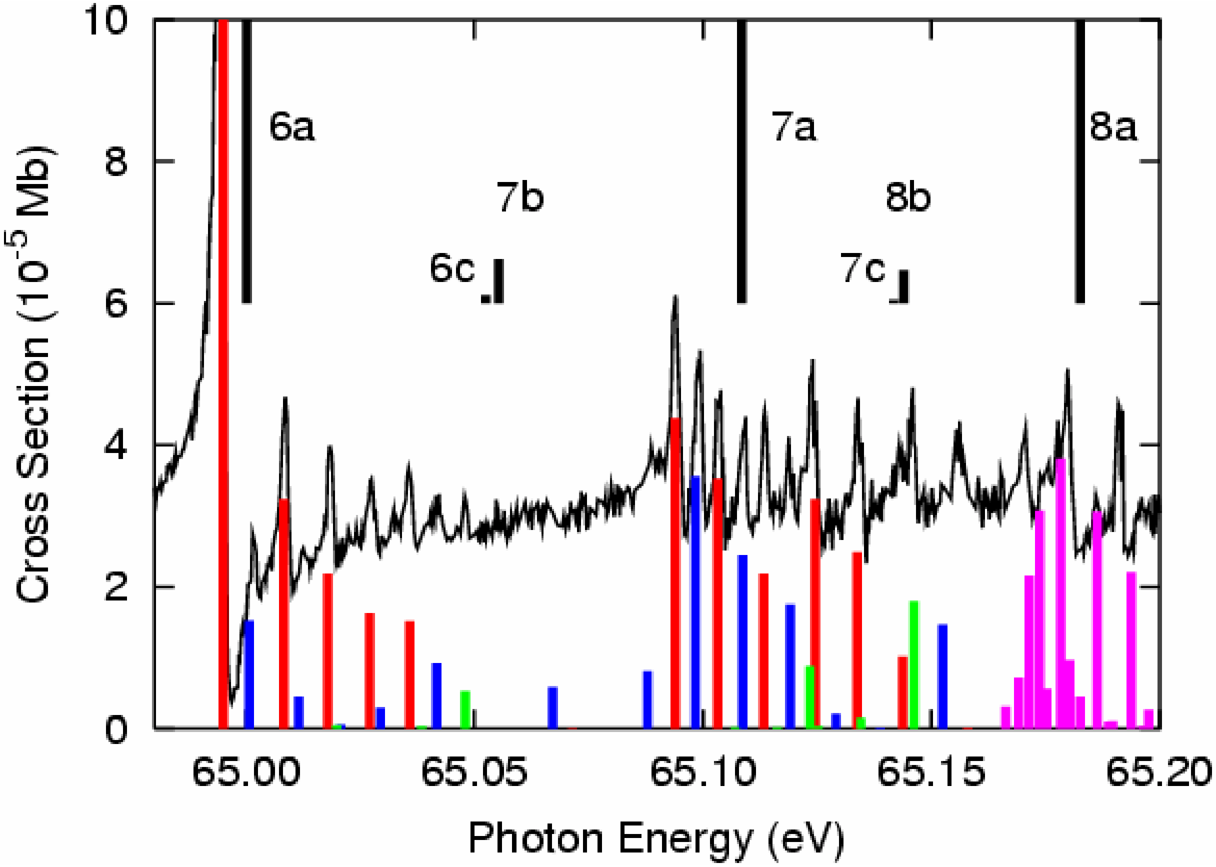
$$He(1s^2\ {}^1S) + h\nu + \mathbf{E} \rightarrow He^{**}(2l\ nl'\ {}^1L)$$

(both dipole allowed and forbidden states)

Application: He** in a Static Field

near 2l6l’, 2l7l’

F=80kV/cm



Expt: J. R. Harries *et. al.*, PRL **90** (2003) 133002.

Theory: Tong and Lin, PRL 92, (2004) 223003

He** in a X-ray and Laser fields (1)

Simulation conditions:

Target: He** $2s^2\ {}^1S$ (7 fs) , $2p^2\ {}^1S$ (140 fs), ${}^1D$ (17 fs)
and $2s2p\ {}^1P$ (17 fs)

X-ray:

pulse duration: 0.2 fs;
center frequency: $1s^2\ {}^1S \rightarrow 2s2p\ {}^1P$
Intensity: 10^{12} W/cm^2

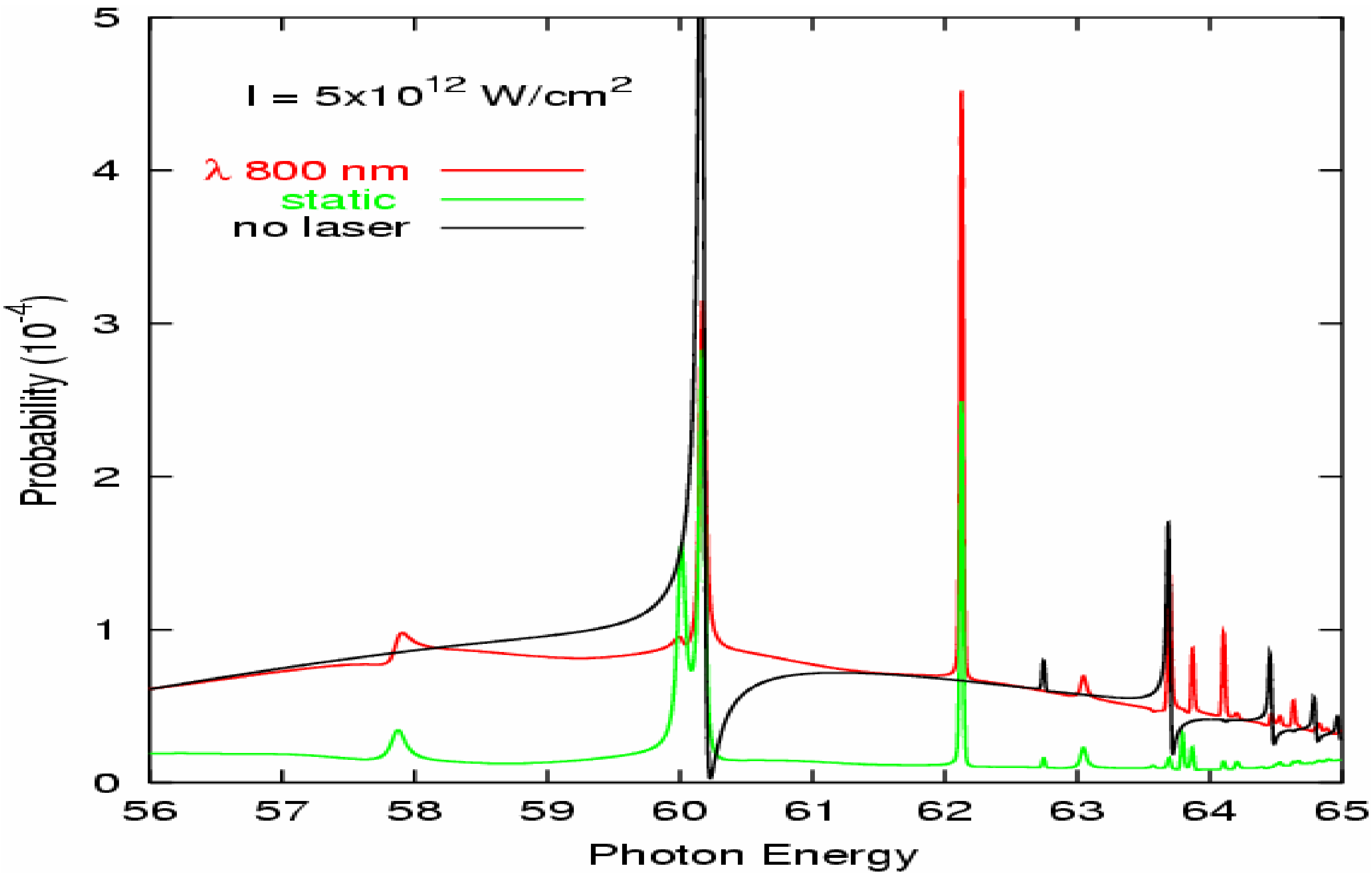
Laser:

pulse duration: 2.0 fs;
center wavelength: 300 nm » 1
Intensity: 10^{12}» 10^{13} W/cm^2
Absolute phase: $\delta = 0$

Note: such a condition can not be reached in the experiment so far,
but it will help us to understand the physics.

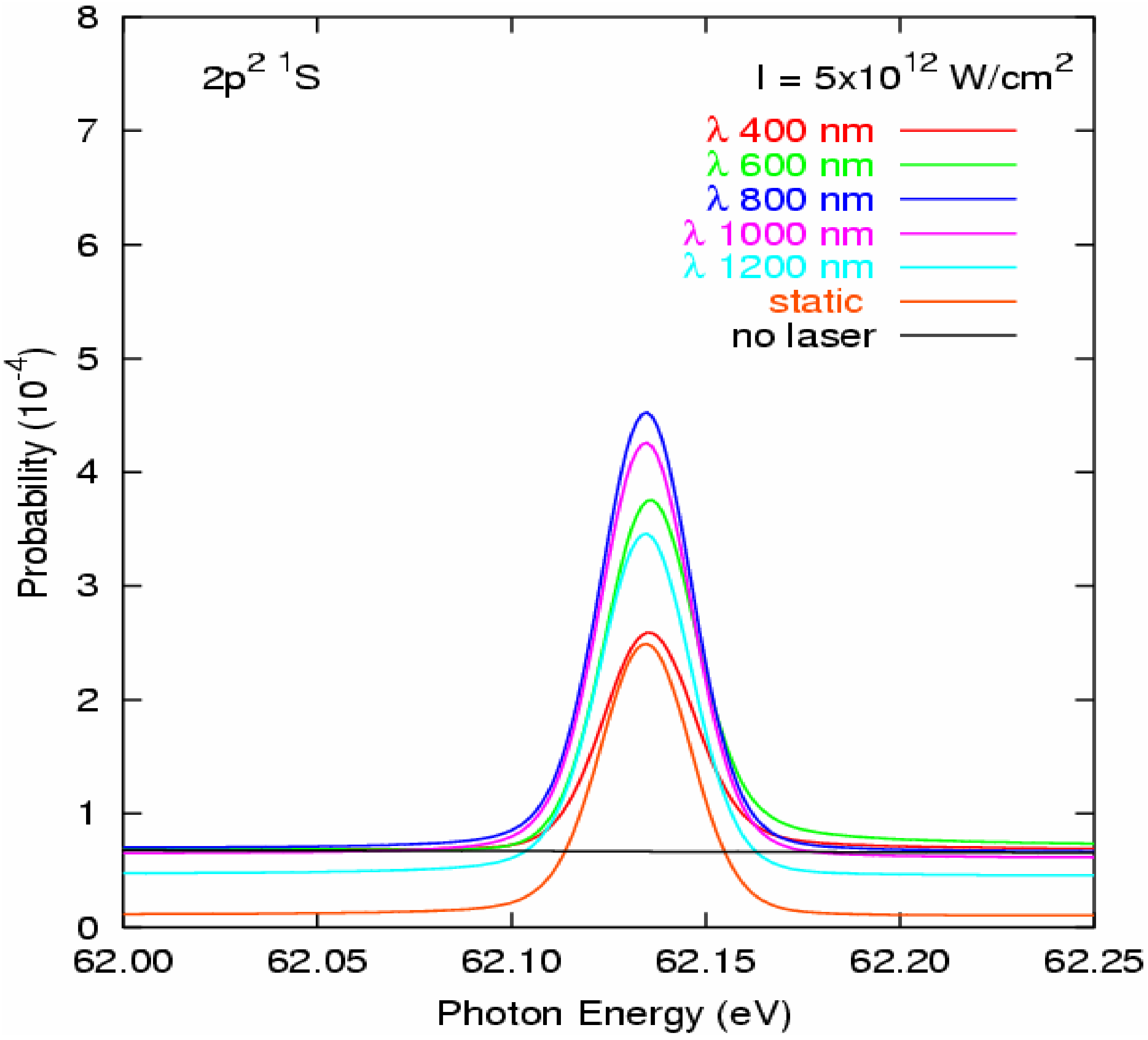
He** Spectra (I)

x-ray 0.2fs
laser 2fs

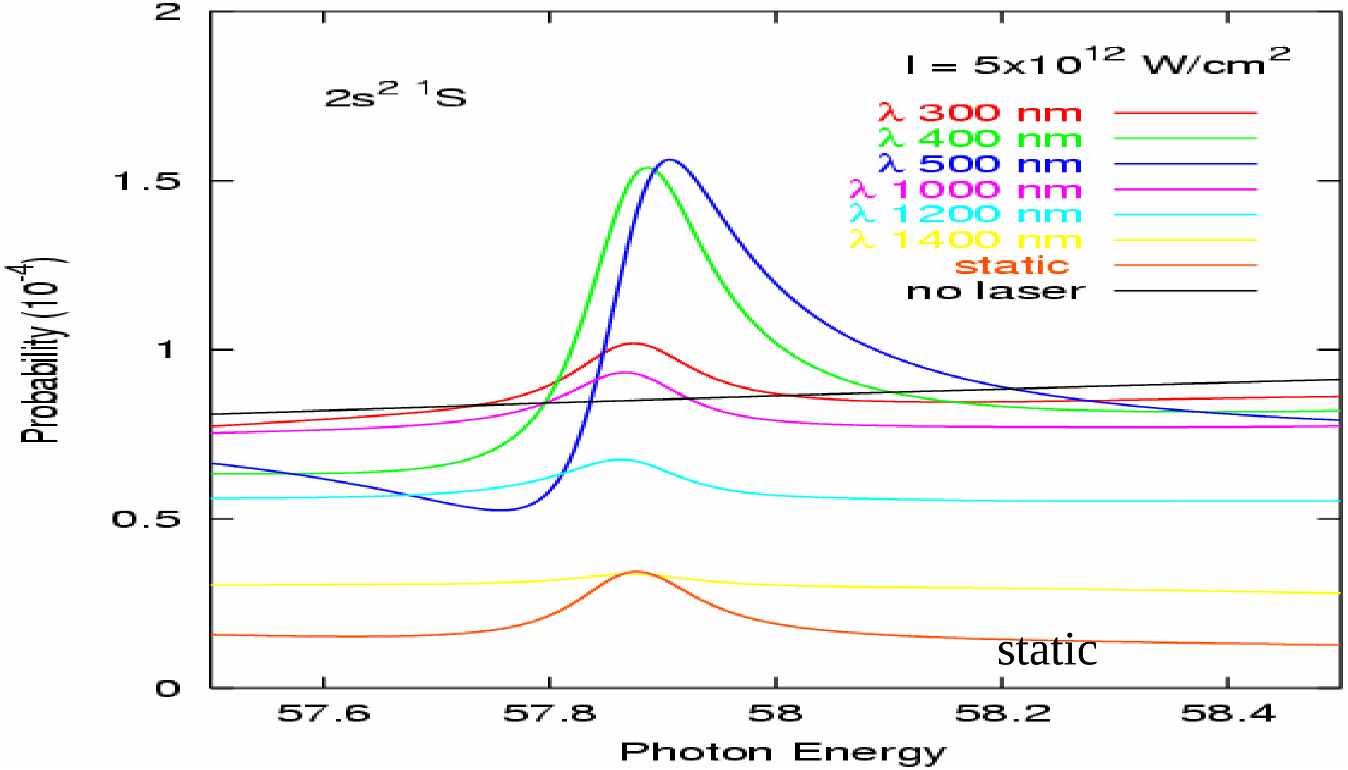


S D,P S

Long lifetime states



Short lifetime states



The **drift velocity will wash out the structure**.
The strengths are lower than the long lifetime states.

He** in a X-ray and Laser fields (2)

Simulation conditions:

Target: He** $2s^2\ {}^1S$ (7 fs) , $2p^2\ {}^1S$ (140 fs), ${}^1D$ (17 fs)
and $2s2p\ {}^1P$ (17 fs)

X-ray:

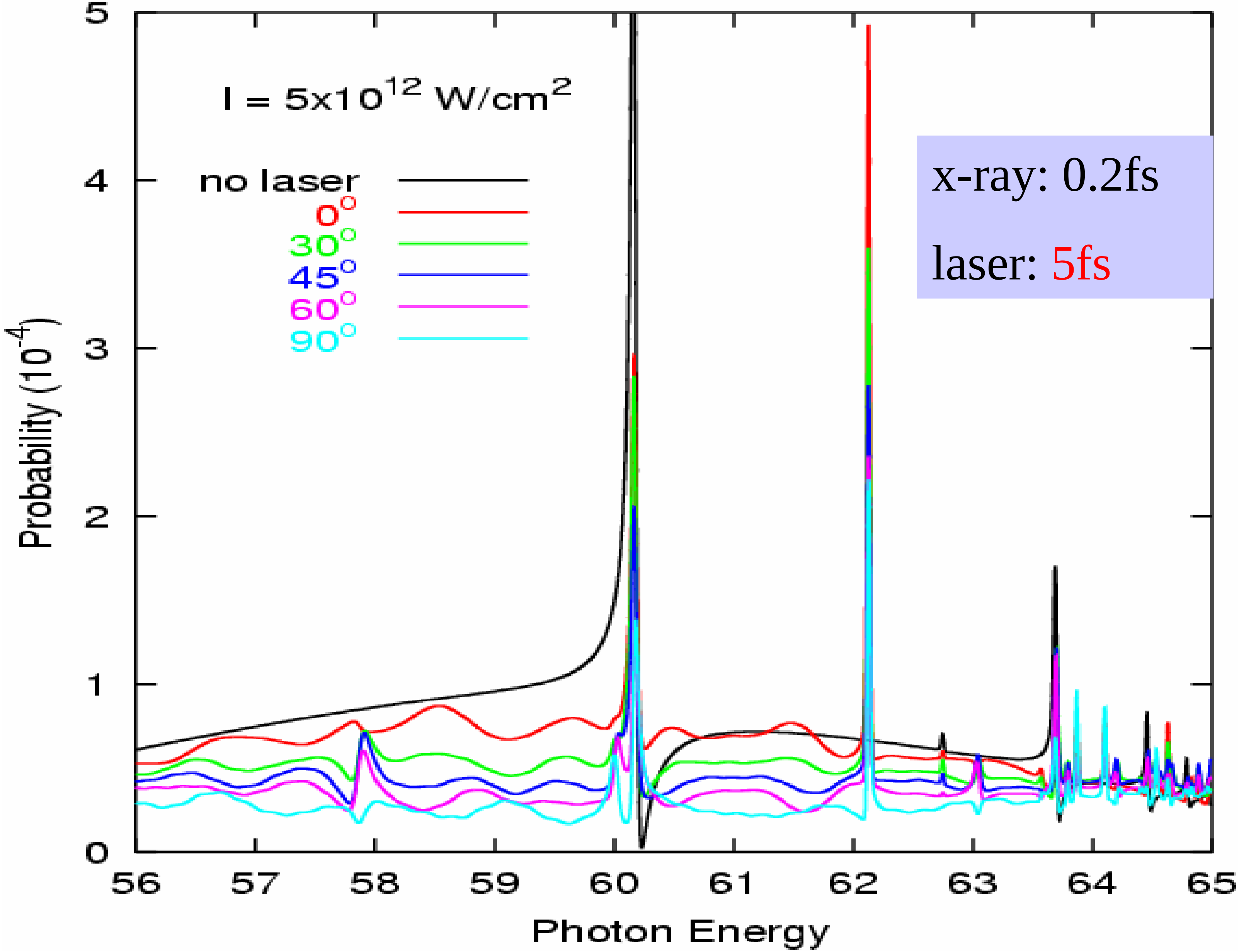
pulse duration: 0.2 fs;
center frequency: $1s^2\ {}^1S \rightarrow 2s2p\ {}^1P$
Intensity: 10^{12} W/cm^2

Laser:

pulse duration: 5.0 fs;
center wavelength: 800 nm
Intensity: 10^{12}» 10^{13} W/cm^2
Absolute phase: 0» $\pi/2$

Note: such a condition can be reached in the experiment so far.

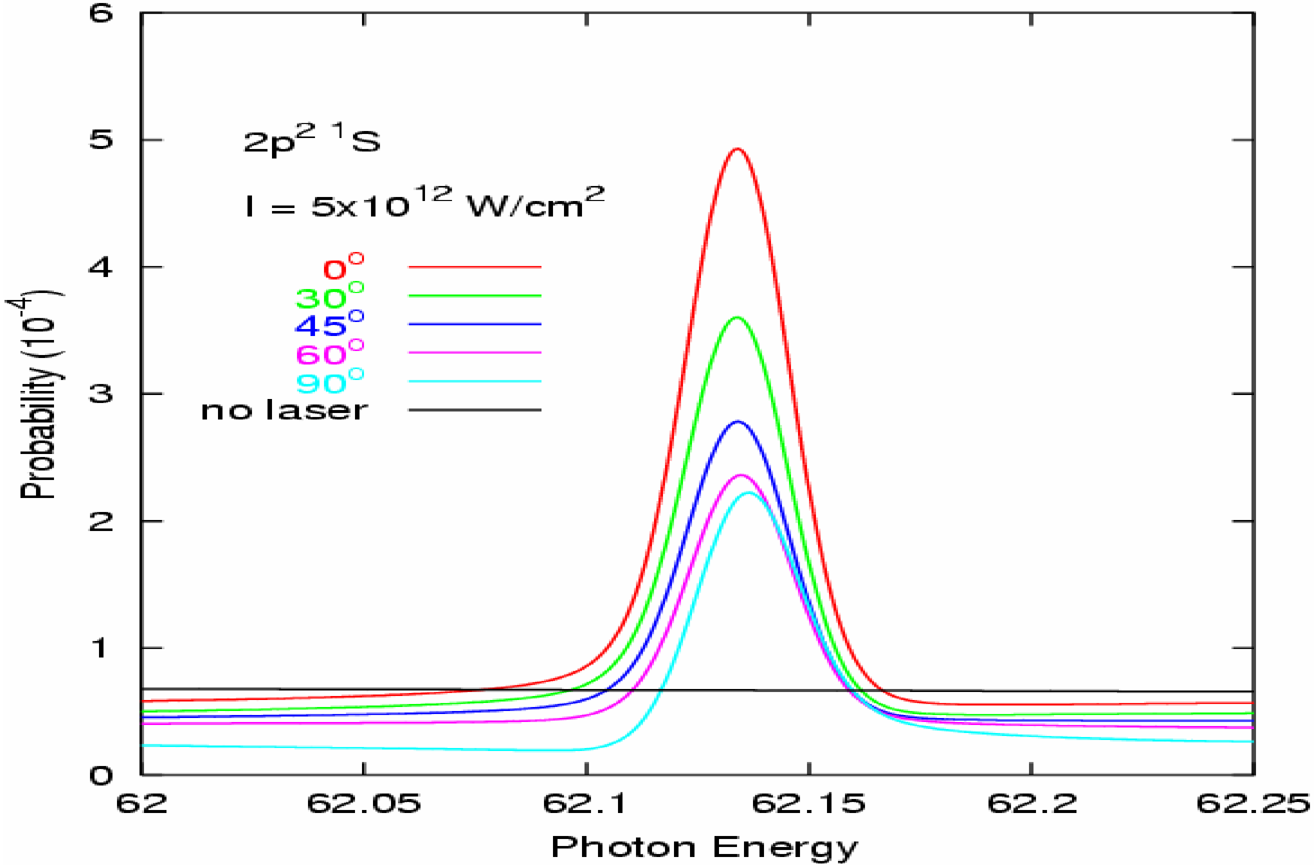
He** Spectra (II)



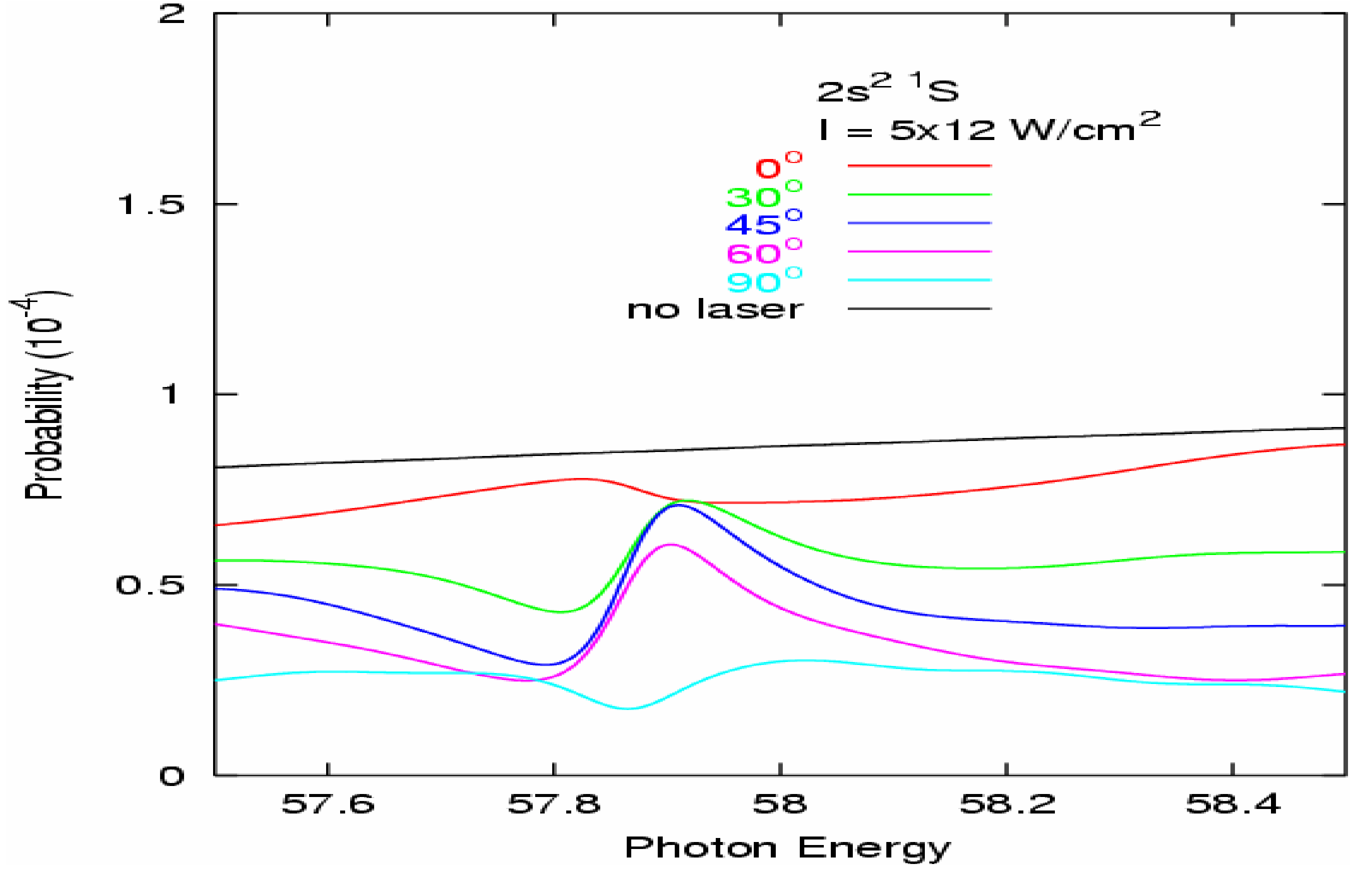
x-ray: 0.2fs

laser: 5fs

Long lifetime states—carrier phase dependence



Short life time states- carrier phase dependence



Extension of the theory to picosecond x-ray pulses?

- Laser-assisted photoabsorption–
  extend Stark field studies to very high electric fields

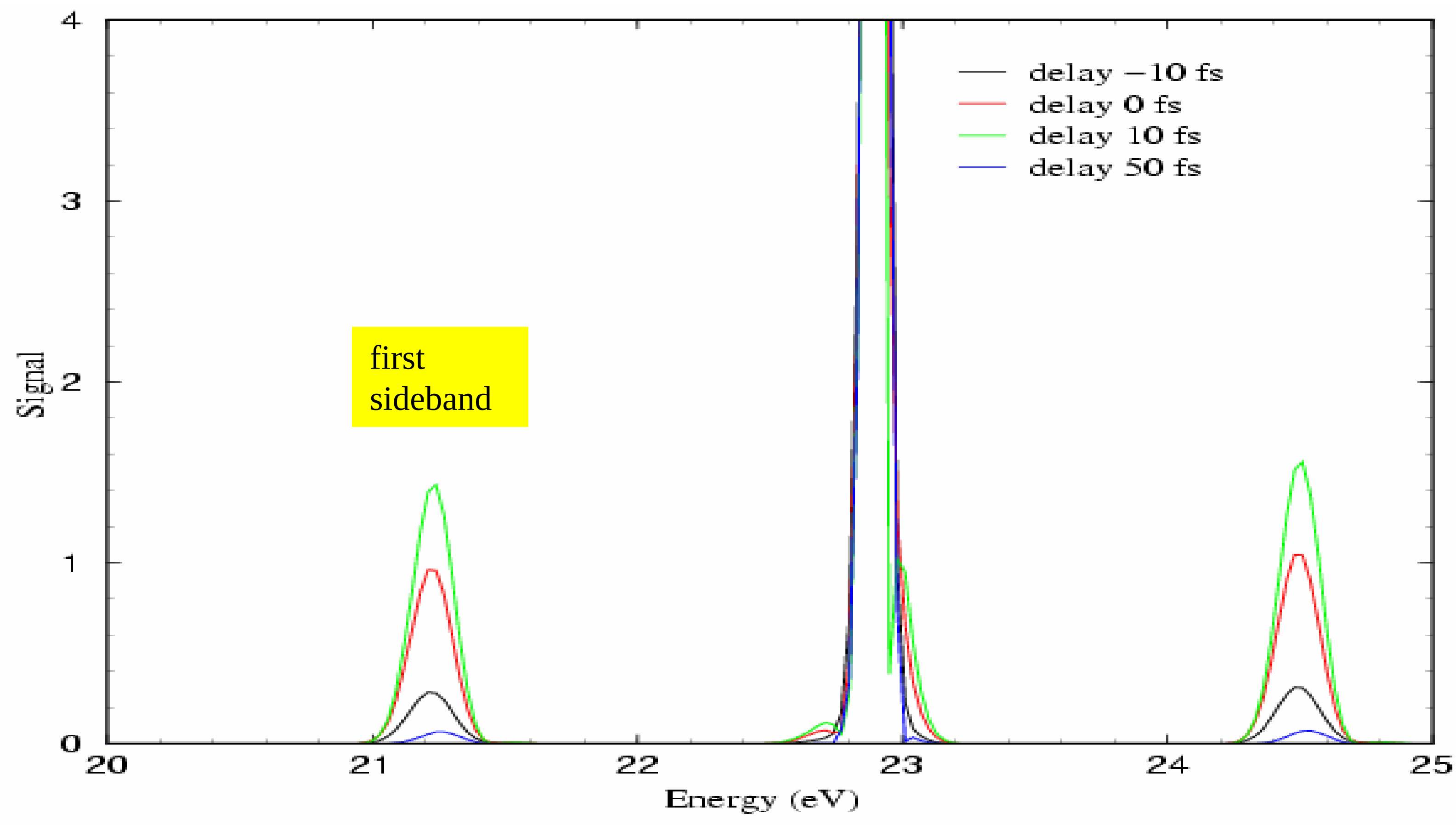
- Laser-x-ray pump-probe experiments to characterize time-structure of x-ray pulses?

Simulated Electron Spectra:

Resonance: at 22.9 eV, 10fs

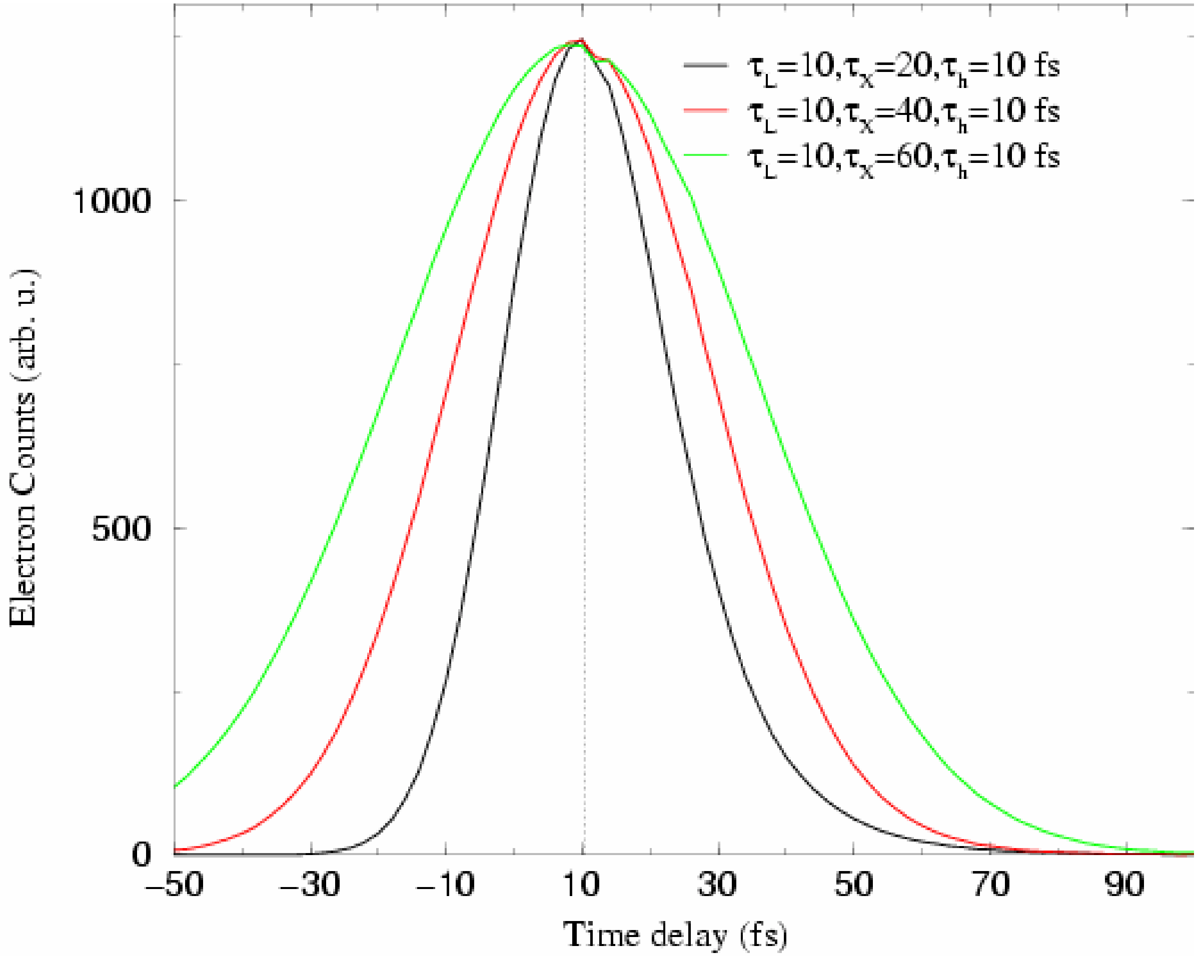
X-ray: 39eV, 20fs pulse, 10^{12} W/cm^2

Laser: 1.65eV, 10fs pulse, 10^{12} W/cm^2



Electron counts within the first sideband versus the time delay between x-ray and laser pulses

Gaussian pulses



X-ray duration dependence

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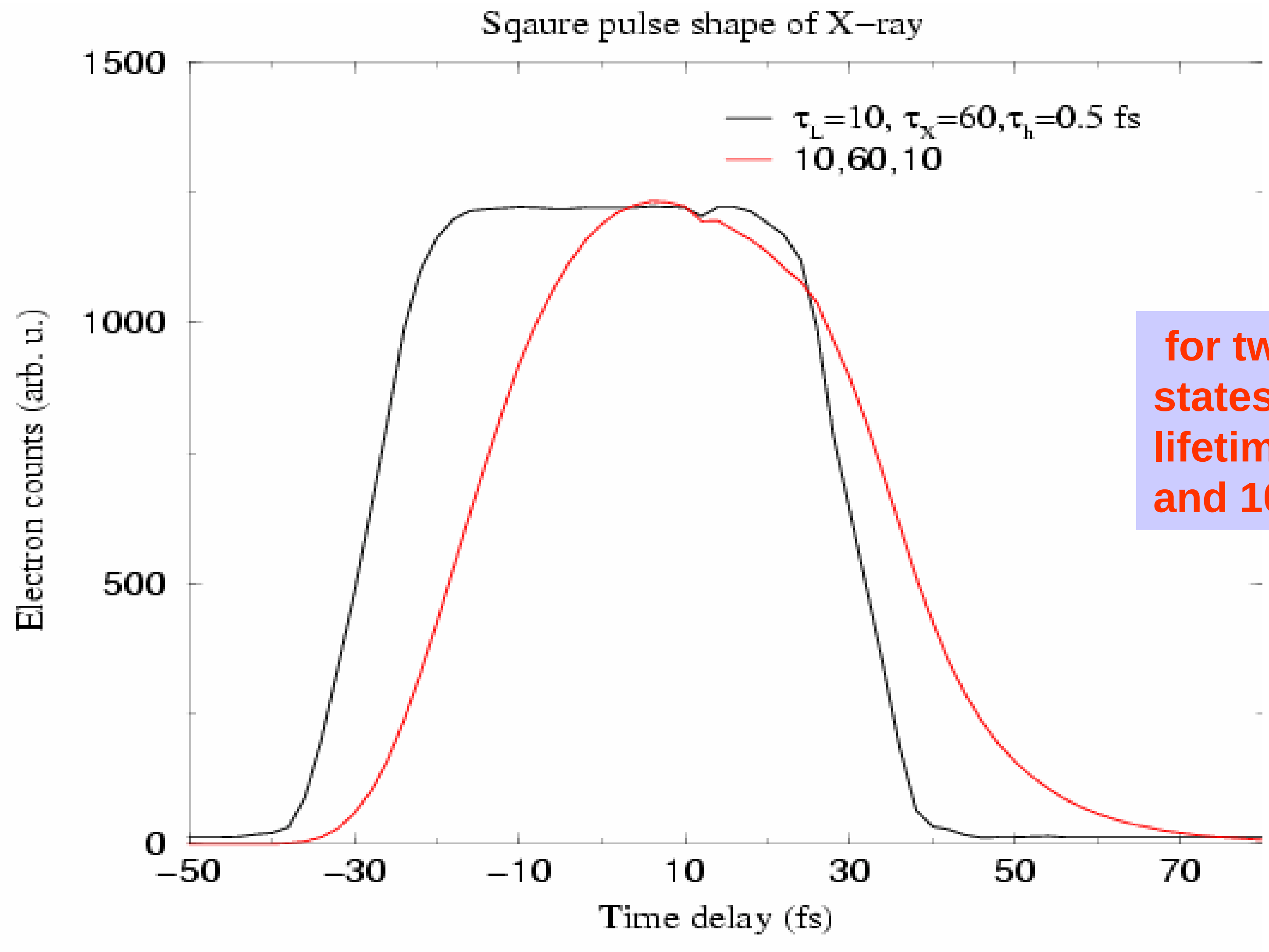
Resonance energy: 22.9 eV, first sideband located in 20.4-22.1eV
laser freq:1.65 eV, intensity 10^{12} W/cm^2
X-ray intensity 10^{12} W/cm^2 with photon energy 39 eV.

Electron counts within the first sideband
Vs the time delay between x-ray and laser pulses

Square pulses



for two Auger states of lifetimes of 0.5fs and 10fs.

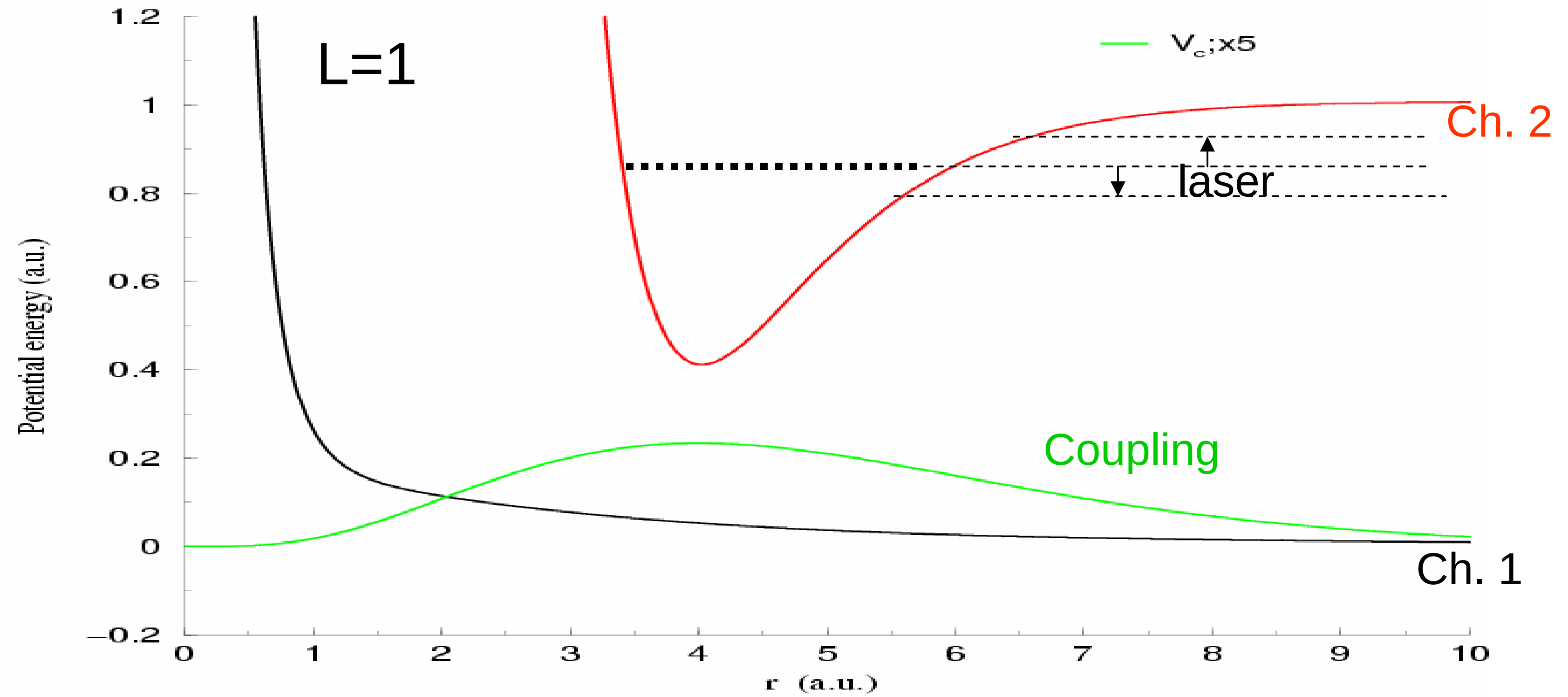
Numerical model

Two-channel TDSE to model two-e- system in a laser field :

$$i\frac{\partial}{\partial t}\begin{pmatrix} F_1 \\ F_2 \end{pmatrix} = \begin{bmatrix} H_1 & V_c \\ V_c & H_2 \end{bmatrix}\begin{pmatrix} F_1 \\ F_2 \end{pmatrix} + \begin{bmatrix} \vec{r}\cdot\vec{E} & \vec{R}_c\cdot\vec{E} \\ \vec{R}_c\cdot\vec{E} & \vec{r}\cdot\vec{E} \end{bmatrix}\begin{pmatrix} F_1 \\ F_2 \end{pmatrix}$$

- Split-Operator propagation method used to solve TDSE
- Two channel continuum constructed by applying scattering wave boundary condition

Feshbach resonance: two-channel potential with coupling

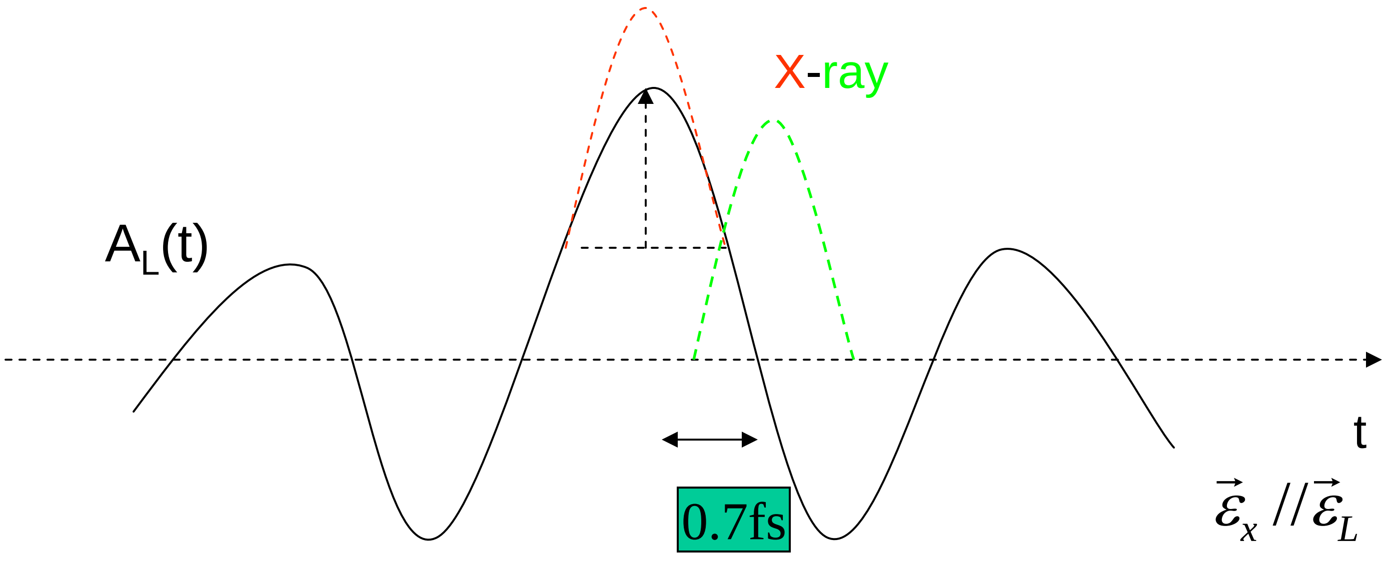


Energy gap 27.21 eV
Resonance 23 eV
Ground state -16.1 eV

Xray pulse: 0.5 fs, 1x10^{12} W/cm^{2}, 38.1 eV
Laser: 10 fs, 2x10^{12} W/cm^{2}. Phase:0 and frequency 0.04 a.u.(1 eV).

$\vec{\varepsilon}_x \,//\, \vec{\varepsilon}_L$

Example of attosecond metrology: Laser-assisted Photoionization

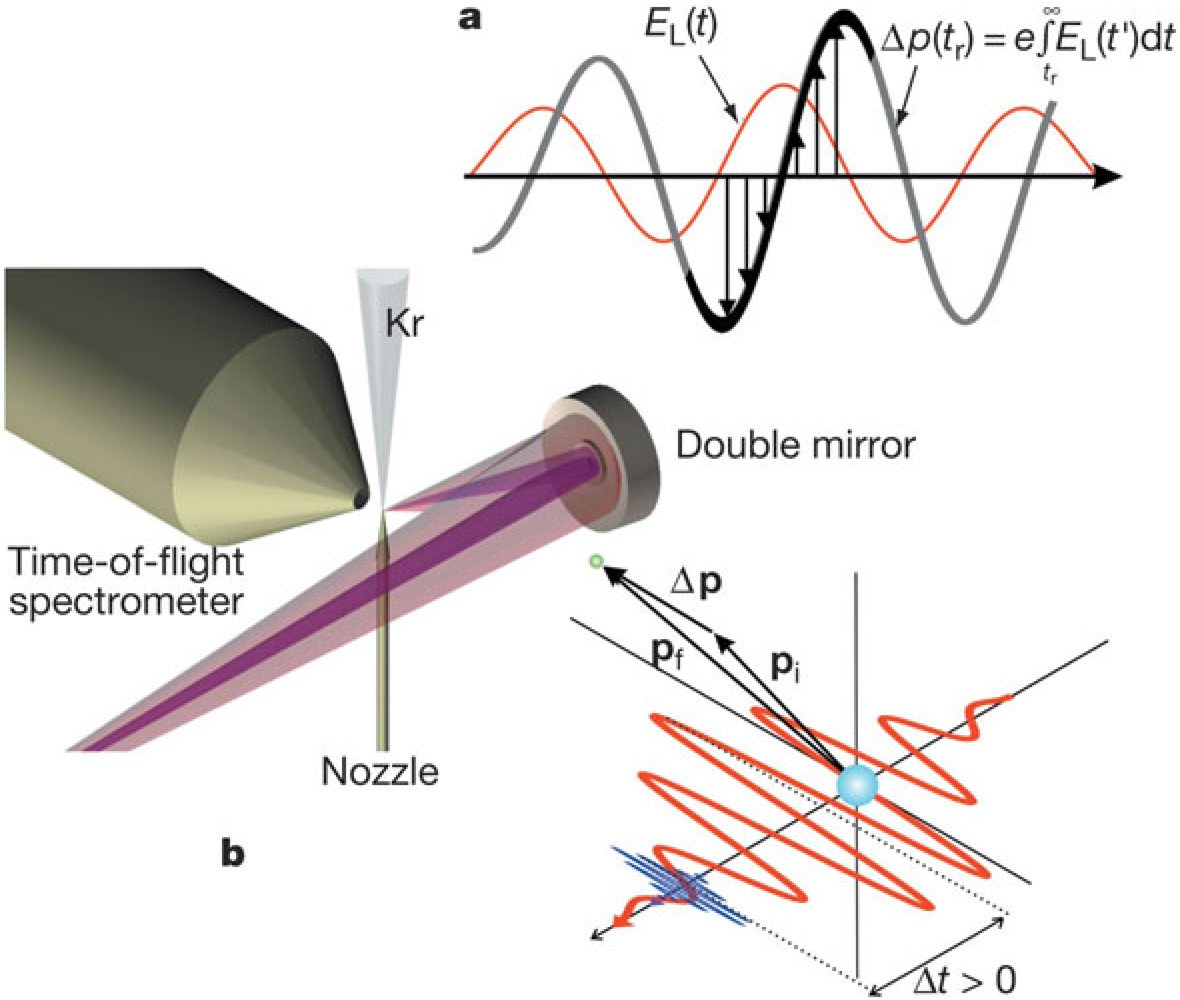


Spectrum:

W/o Laser Delay=0 Delay=T/4

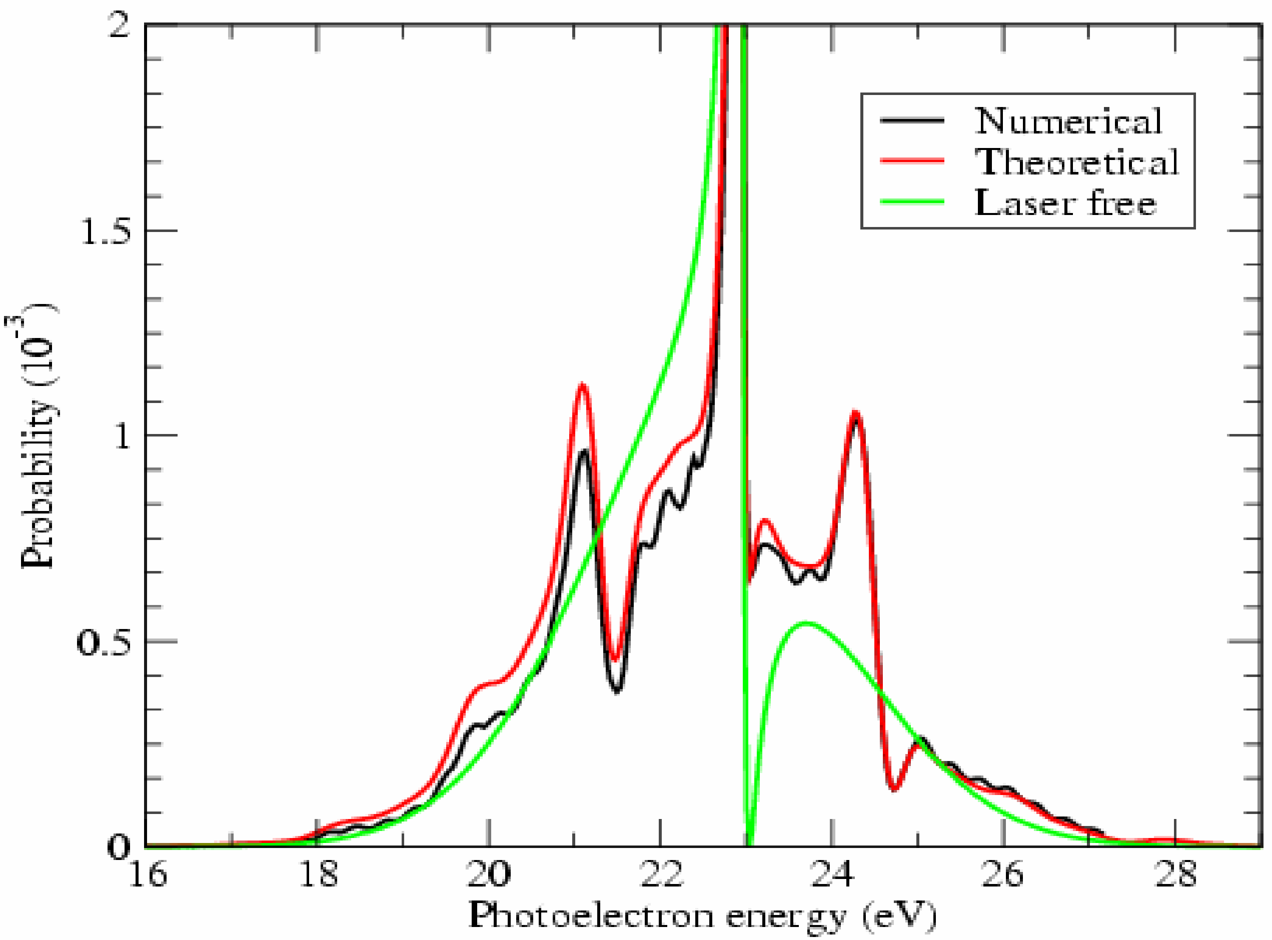
photoelectrons steered by the laser field

Kitzler et al, PRL88, 173904



Drescher et al, Nature 419, 803

Angle-Integrated spectra (delay =2.5fs)



Xray: 0.5 fs, 1×10^{12} W/cm^2, 39 eV

Laser: 5 fs, 1×10^{12} W/cm^2. Phase:0 and 1.65 eV (~759nm, T=2.5fs).